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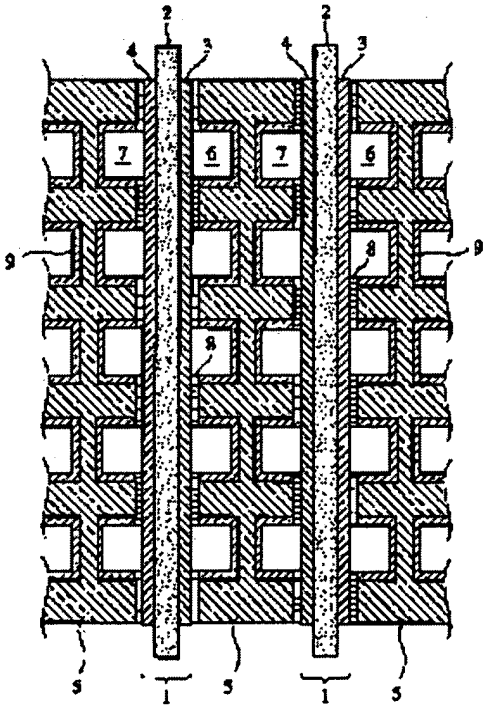
(54) ON-VEHICLE FUEL CELL

(57)Abstract:
PROBLEM TO BE SOLVED: To provide an on-vehicle fuel cell using separators of light weight and low-cost, and equipped with a high current collecting performance and a long-term stability.
SOLUTION: An on-vehicle fuel cell includes laminated unit cells 1 each composed of a solid electrolyte 2 and an anode 3 and cathode 4 installed on both sides thereof, and the anode 3 and cathode 4 are in contact with separators 5 and connected with an external circuit, not illustrated, through the two ends of the laminate. The base material of the separators 5 consists of a compratively light metal having good anti-corrosiveness and a high resistance against oxidation, and its both surfaces are provided with grooves 6 to serve as passage for reaction gas. A fuel gas is supplied to the passage bound by the grooves 6 and anode 3 while an oxidizer gas is supplied to the passage bounded by other grooves 7 in the separators 5 and the cathode 4. The separators 5 are made of Al, Ti, or alloy thereof, and it is preferable that protection films 9 are formed at least on the inner walls of these grooves 6 and 7 for enhancement of the anti-corrosiveness for hot water (steam) and resistance against oxidation.



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* NOTICES *

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2.**** shows the word which can not be translated.
3.In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] About the fuel cell generated by the electrochemical reaction of fuel gas, such as hydrogen gas, and oxygen gas, this invention is lightweight so that it may be suitable for especially mount, it has the high current collection engine performance and long term stability, and relates to the fuel cell of low cost.
[0002]
[Description of the Prior Art] A fuel cell comes to carry out the laminating of the unit cell (cel) which generally consists of the anode and cathode which were formed in an electrolyte layer and its both sides through a separator. Between the anodes and cathodes in each separator and its both sides, the path of reactant gas is prepared, respectively.
[0003] Reactant gas consists of fuel gas and oxidant gas, fuel gas is supplied to the anode side path of a separator, and oxidant gas is supplied to the cathode side path of a separator. An electron (electrical energy) generates with progress of the electrochemical reaction of such reactant gas, and this is taken out to an external circuit with lead wire. In the fuel cell, since there are no exhaust gas problems, such as NOx, CO2, and soot, like the usual engine while it is possible to transform reaction energy into electrical energy at very high effectiveness, it is expected as a next-generation power plant for electric vehicles.
[0004] A property required as a fuel cell for mount is efficient generation-of-electrical-energy engine-performance, endurance [from which an output is obtained stably for a long period of time], lightweight-izing, and low cost-ization etc. Fundamentally, the conventional fuel cell for mount developed as what has such a property consists of layered products which carried out the laminating of the unit cell (cel) 1 which consists of the anode 3 and cathode 4 which were formed in an electrolyte membrane 2 and its both sides through the separator 5, as shown in drawing 3 . The slot 6 for fuel gas supply is formed in the anode side front face of a separator 5, and the slot 7 for oxidant gas supply is formed in the cathode side front face. The separator 5 which touches the anode 3 and cathode 4 which were installed in the both sides of each unit cell 1 is conductivity, and is connected to the external circuit (not shown) from layered product ends.
[0005] The separator 5 which has the above-mentioned structure must have good corrosion resistance and good oxidation resistance in order to contact the hot (near 100 **) steam which generates the inside of the slot 6 and 7 by the reaction of fuel gas and oxidant gas while reactant gas flows. Moreover, in order to lead efficiently the current acquired by the electrochemical reaction to the unit cell 1, it is necessary to make the bulk resistor of a separator 5 low and to make low current collection resistance (contact resistance) in the contact surface with an electrode. Furthermore in consideration of mass production nature, the ingredient which is easy to process it must be used.
[0006] Although the separator with which the above-mentioned property is demanded is mainly a product made from a graphite, it has the problem that the separator made from a graphite has the high processing cost of the slot for reactant gas paths, and that of the material cost itself is high now.

Moreover, although binding material must be added to carry out shaping solidification using graphite powder, this binding material reduces the current collection engine performance of an electrode, and has the problem of increasing a bulk resistor. Furthermore, the graphite itself also has the problem that the dependability over a mechanical strength, especially an impact is low.
[0007] Development of the separator using a metallic material is also performed in view of such a situation. For example, JP,2-247977,A has reported the separator which comes to carry out thermal spraying or paste coating of aluminum to a stainless steel base material. Moreover, JP,8-236127,A has reported the separator which comes to carry out nickel plating to the anode plate side of the base material made from stainless steel. However, when these separators are used, the weight of a fuel cell will increase and current collection performance degradation preventing will become inadequate. Further stainless wet etching and press working of sheet metal also have the problem of being cost high.
[0008] Therefore, the object of this invention is providing utilization called low cost with the fuel cell for mount possessing the separator which fulfills indispensable conditions simultaneously while it has the high current collection engine performance and long term stability and is lightweight.
[0009]
[Means for Solving the Problem] While this invention person used aluminum, Ti, or these alloys for the base material of a separator wholeheartedly in view of the above-mentioned technical problem as a result of research, when the precise conductive coat which has corrosion resistance was formed in the electrode contact surface,-izing of the fuel cell could be carried out [lightweight], corrosion resistance improved, it discovered that good current collection nature could be secured and long-term stability could be secured, and this invention was completed.
[0010] that is , the fuel cell of this invention come to carry out the laminating of the unit cell which formed the anode and the cathode in both sides of the solid electrolyte which be a proton conductor through a separator , and the path of reactant gas be prepare between said separator and said anode , and said cathode , respectively , and said separator consist of a base material which use a metal as a principal component , and be characterize by form the precise conductive coat of said separator base material which have corrosion resistance in the electrode contact surface at least .
[0011] As for the base material of the viewpoint of conductivity, corrosion resistance, oxidation resistance, and lightweight-izing to a separator, it is desirable to form with aluminum, Ti, or these alloys. Moreover, as for the construction material of the conductive coat formed in the base material front face of a separator, it is desirable that they are Au, Ag, Pt, Pd, nickel, Cr, W or these alloys, carbon, or conductive carbide.
[0012] When there is a part which is not covered with a conductive coat on a separator front face, as for the part, covering with a protective coat is desirable. As construction material of a protective coat, oxides, such as aluminum, Ti, and Cr, a nitride, etc. are desirable.
[0013]
[Embodiment of the Invention] This invention is explained to a detail below with reference to an accompanying drawing. In addition, since the basic structure of the fuel cell by one example of this invention shown in drawing 1 is the same as what is shown in drawing 3 , the same reference number has been given to the corresponding part.
[0014] [1] It comes to carry out the laminating of the unit cell 1 which the fuel cell of the example shown in unit cell drawing 1 consisted of with the anode 3 and cathode 4 which were formed in a solid electrolyte 2 and its both sides. The anode 3 and the cathode 4 touch the separator 5, respectively, and are connected to the external circuit (not shown) from layered product ends.
[0015] (1) As a solid electrolyte solid electrolyte 2, it is H+ generated by the electrochemical reaction. Although any ingredients can be used if it is the matter with the conductivity of ion, cation exchange membrane including the viewpoint of thinning to Nafion (Nafion) etc. is desirable.
[0016] (2) As for the electrode anode 3 and a cathode 4, it is desirable to come to apply the paste containing the powder and water repellents of noble metals, such as carbon black and Pt, to the solid electrolyte plate 2, and its two electrodes are good with the same construction material. Carbon black is 50 - 93 % of the weight, an exotic powdered metal is 2 - 50 % of the weight, and, as for the presentation

of each electrode, it is desirable that water repellent is 5 - 30 % of the weight. Moreover, as for the thickness of each electrode, it is desirable that it is 1-20 micrometers.
[0017] [2] Separator (1) It is comparatively lightweight, and corrosion resistance and oxidation resistance consist of a high metallic material, and aluminum, Ti, or these alloys of the base material of the base material separator 5 are specifically desirable. As for the thickness of the plate for separators, it is desirable that it is 1-5mm, and especially its about 3mm is desirable.
[0018] (2) The base material of the slot separator 5 has the slot 6 for reactant gas paths to both sides. Fuel gas is supplied to the path formed with the slot 6 and anode 3 of a separator 5, and oxidant gas is supplied to the path formed by the slot 7 and cathode 4 of a separator 5. Slots 6 and 7 can be formed by performing press working of sheet metal, wet etching, etc. to the plate for separators. The depth of each slots 6 and 7 is usually 0.5-1.5mm. Carrying out is desirable, and the configuration of slots 6 and 7 has small reactant gas ventilation resistance, and it is desirable to set up so that generation efficiency may become high.
[0019] The shape of a quirk of a separator 5 is not limited to what is shown in drawing 1 , but just forms the path for reactant gas in the part which touches an anode 3 and a cathode 4. For example, as shown in drawing 2 , an aluminum plate etc. may be made to transform in the shape of corrugated one, and slots 6 and 7 may be formed in both sides by turns. In addition, although only the conductive coat 8 is formed in the example of drawing 2 , of course, a protective coat may be formed in fields other than an electrode contact part.
[0020] (2) In order to make low contact resistance with conductive coat each electrodes 3 and 4, form the conductive coat 8 in the part of the separator 5 which contacts each electrodes 3 and 4 at least. As construction material of the conductive coat 8, it is desirable to use **Au, Ag, Pt, Pd, nickel, Cr(s) and W or these alloys, ** carbon, or ** conductivity carbide. The thickness of the conductive coat 8 is good at about 0.1-10 micrometers.
[0021] ** a case -- Au, Ag, Pt, Pd, etc. -- contact resistance of corrosion resistance is [a noble-metals system coat] very good low. Moreover, low cost-ization is more realizable if the coat of nickel, Cr, or W is used. These metallic films can be formed by plating or PVD.
[0022] ** a case -- CVD -- depending -- a graphite -- the film -- or -- DLC -- the film (diamond-like carbon film) -- etc. -- being desirable . Moreover, what added water repellent may be applied to graphite powder. Since the electrode which is formed in the electrolyte plate 2 which consists of an ion exchange membrane in the case of a carbon coat becomes carbon black from what added Pt of a minute amount, contact concordance is good by the same carbon systems.
[0023] Moreover, in **, it is desirable to use the coat of conductive carbide, such as silicon carbide, carbonization niobium, and tungsten carbide. Since it contact resistance is not only small, but has good corrosion resistance and good oxidation resistance, a carbide coat acts also as a protective coat of a separator 5.
[0024] (3) In order to raise the corrosion resistance and the oxidation resistance over hot water (steam) of the separator 5 which consists of protective coats aluminum and Ti or these alloys, it is desirable to form a protective coat 9 in the wall of slots 6 and 7 at least. The thickness of a protective coat 9 is good at about 1-10 micrometers. There are the approach of oxidizing ** separator base material and the approach of carrying out the laminating of the coat of ** corrosion resistance matter in formation of a protective coat 9.
[0025] ** It is desirable to form an alumite coat in the case of an approach (for example, anodic oxidation of aluminum). It considers as a precise boehmite film (gamma-aluminum 2 O 3 and H 2 O) by forming a gamma-alumina coat in a base material front face by electrolysis at an anode oxidation method, using water solutions, such as oxalic acid, a sulfuric acid, and a chromic acid, as the electrolytic solution, and subsequently processing with a boiling water or a heating steam. Once a boehmite film is formed, under the usual actuation conditions, the separator 5 is dramatically stable.
[0026] Moreover, coats, such as the case of the approach of **, for example, an alumina, and a titania, can be formed with a spraying process, the sputtering method, the ion plating method, a CVD method, etc.

[0027]
[Example] Although the following examples explain this invention to a detail further, this invention is not limited to them.
[0028] Press working of sheet metal is performed to each of the aluminum plate (3mmx150 mmx150 mm) of 12 examples, and it is a depth of 1.0mm. And width of face of 3.0mm The separator base material which has a slot for reactant gas paths was produced. The alumite coat was formed by drying, after electrolysis in an oxalic acid water solution performs anodizing and being immersed subsequently to ebullition underwater to each separator base material for about 30 minutes. Subsequently, in order to improve the display flatness of the electrode contact surface of each separator, it washed, after carrying out lap polish of the electrode contact surface. Thereby, the alumite coat of the electrode contact surface was removed. Next, 5mTorr Sputtering of the Pd-Au alloy was carried out to the electrode contact surface of a separator base material, having used substrate temperature as 200 ** in the pure argon gas ambient atmosphere, and the separator of the layer system shown in drawing 1 was obtained. The conductive coat which consists of a Pd-Au alloy was 1 micrometer in thickness.
[0029] Moreover, carbon black 100 After adding the Pt paste (Pt:90 % of the weight) 15 weight section in the weight section, the Teflon particle (mean particle diameter: 0.2 mum) 15 weight section was added as water repellent, and the paste for electrodes was produced. This was applied to the carbon cross paper and it dried. The unit cell of the structure shown in drawing 1 was produced by pasting up the carbon cross paper which applied the paste for electrodes on the Nafion (Nafion) film (cation exchange membrane), and inserting into it with the above-mentioned separator of two sheets. The path for fuel gas was formed with one slot and anode of a separator, and the path for oxidant gas was formed by the slot and cathode of a separator of another side.
[0030] In the room temperature, the hydrogen gas (the rate of flow: 40 ml/min) of 100 % was supplied for the unit cell to the path for fuel gas by the side of an anode, and it generated electricity by supplying dry air (the rate of flow: 100 ml/min) to the path for oxidant gas by the side of a cathode. The output voltage and the short-term output drift stability between two electrodes were measured. A result is shown in a table 1.
[0031] The separator which consists only of a graphite base material instead of the separator of one to example of comparison 3 example 1, respectively (example 1 of a comparison), The separator which consists only of a stainless-steel (SUS 304) base material (example 2 of a comparison), And the separator (example 3 of a comparison) which consists only of an aluminum base material was used, the unit cell was produced by the same approach as an example 1, and it generated electricity by supplying hydrogen gas and dry air to each path for gas on the same conditions as an example 1. The output voltage and the short-term output drift stability between two electrodes were measured. A result is shown in a table 1.
[0032]
Table 1 Example No. Configuration of a separator Output voltage (V) Drift stability example 1 aluminum base material + alumite Coat +Pd-Au Alloy film 0.82 Example 1 of a good comparison Graphite simple substance 0.83 Example 2 of a good comparison SUS 304 Simple substance 0.57 It is the example 3 of a defect comparison a little. aluminum simple substance 0.49 Defect [0033] Although it is equivalent to the unit cell (example 1 of a comparison) which possesses the separator of the product [cell / of an example 1 / unit] made from a graphite about the output voltage and drift stability as cell engine performance so that clearly from a table 1, it excels as compared with the unit cell (example 3 of a comparison) possessing the separator which consists of the unit cell (example 2 of a comparison) and aluminum simple substance possessing the separator which consists of a stainless-steel simple substance. However, since the unit cell of an example 1 is remarkably excellent from a viewpoint of cost and mechanical dependability as compared with the thing of the example 1 of a comparison, the predominance of the fuel cell of this invention to elegance is conventionally clear.
[0034] The unit cell was produced like the example 1 except having formed the silicon carbide coat (conductive coat) in the whole base material front face by the plasma-CVD method, without forming an alumite coat in the separator which uses example 2 aluminum as a base material. The thickness of a

silicon carbide coat could be about 0.5-5 micrometers by adjusting membrane formation time amount. Thus, the output voltage and the short-term output drift stability between two electrodes were measured by supplying hydrogen gas and dry air to the path for reactant gas of the unit cell possessing the produced separator on the same conditions as an example 1. A result is shown in a table 2.
[0035]
Table 2 silicon-carbide thickness Cel output voltage Drift (micrometer) (V) Stability 0.46 0.53 Defect 0.98 0.64 He is a defect 1.48 a little. 0.79 It is good 1.95 a little. 0.81 Fitness 3.20 0.83 Fitness 4.11 0.82 Fitness 5.19 0.80 Fitness [0036] It is about 1.5 so that clearly from a table 2. By considering as the thickness more than mum shows that the good cell engine performance is obtained. If the coat of carbide like silicon carbide is formed in the separator of aluminum as a conductive coat, the cell engine performance conventionally equivalent to elegance can be obtained. When cost and mechanical dependability are taken into consideration, the fuel cell of this example is also known by having a big predominance to elegance conventionally like an example 1.
[0037]
[Effect of the Invention] Large lightweight-ization can be attained by using metallic materials, such as aluminum and Ti, for a separator, and since mass production nature is high compared with the conventional separator made from a graphite, processing cost can be reduced. Moreover, improvement in cost reduction and dependability can be attained, holding the cell engine performance to a conventional graphite separator and conventional equivalent extent by forming the precise conductive coat which has corrosion resistance in the front face of the separator base material which consists of these metallic materials. The fuel cell which has such a description is dramatically promising as a next-generation driving source for automobiles.